NITROGEN ISOTOPIC ANOMALIES IN A HYDROUS INTERPLANETARY DUST PARTICLE. J.B. Smith, Z.R. Dai, P.K. Weber, G.A. Graham, I.D. Hutcheon, S. Bajt, H. Ishii, J.P. Bradley. Institute for Geophysics and Planetary Physics, Lawrence Livermore National Laboratory, PO Box 808, L-231, Livermore, CA 94550, USA (smith371@llnl.gov).

Introduction: Interplanetary dust particles (IDPs) collected in the stratosphere are the fine-grained end member (5 - 50 µm in size) of the meteoritic material available for investigation in the laboratory [1]. IDPs are derived from either cometary or asteroidal sources Some IDPs contain cosmically primitive materials with isotopic signatures reflecting presolar origins, e.g. [6, 7]. Recent detailed studies using the NanoSIMS have shown there is a wide variation of isotopic signatures within individual IDPs; grains with a presolar signature have been observed surrounded by material with a solar isotopic composition [7]. The majority of IDPs studied have been anhydrous [6, 8, We report here results from integrated NanoSIMS/FIB/TEM/Synchrotron IR studies of a hydrous IDP, focused on understanding the correlations between the isotopic, mineralogical and chemical compositions of IDPs.

NanoSIMS Analysis: A large 42 x 34 µm IDP (L2047 D23) was pressed into high purity gold for analysis with the Cameca NanoSIMS 50 ion microprobe at Lawrence Livermore National Laboratory (Fig. 1a). A ~1.5 pA, 16 keV ¹³³Cs⁺ primary ion beam focused into a ~100 nm spot was rastered over a 30 x 30 micron area, at 512 by 512 pixels with a dwell time of 5 ms/pixel. Secondary ion intensities for ¹⁶O⁻, ¹⁷O⁻, ¹⁸O⁻, ¹²C¹⁴N⁻, ¹²C¹⁵N⁻ were collected simultaneously in multi-collection mode, together with secondary electrons. A mass resolving power of ~6500 was used to separate ${}^{13}C_2$ from ${}^{12}C^{14}N$, and ${}^{12}C^{15}N$ from ${}^{13}C^{14}N$. The isotope imaging measurements consist of 30 scans over the area. The data are corrected for instrumental mass-dependent fractionation by normalizing the N-isotopic ratios to NIST potassium nitrate 8558 embedded in graphite; the magnitude of instrumental fractionation is <1%.

The data from L2047 D23 were processed as quantitative isotopic ratio images. Using a variety of criteria to define different regions of interest (ROIs), the full range of ¹⁴N/¹⁵N ratios in the particle can be delineated and ROIs can be identified with highly unusual ¹⁴N/¹⁵N ratios. The secondary electron image of the IDP together with a ¹⁴N/¹⁵N isotope ratio image of the anomalous grain are shown in Fig. 1.

Mineralogical Context: Electron transparent sections were extracted from the pressed IDP using a focused ion beam (FIB) recovery method [10]. These

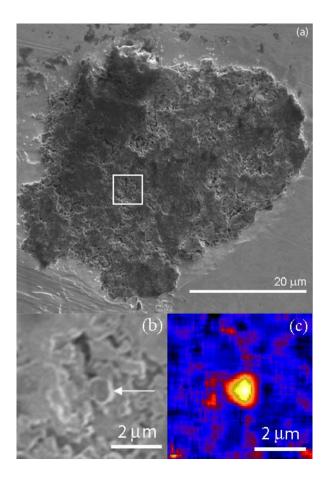


Fig 1: L2047 D23. (a) secondary electron (SE) image of the whole particle. (b) Zoomed view of the anomalous particle (arrowed) within the IDP. (c) False color ¹⁴N/¹⁵N isotope ratio image of the anomalous particle.

sections were characterized using a CM300 FEGTEM and a Tecnai G2 F20 UT (S)TEM with EDS and EELS. From the FIB sections L2047 D23 appears to be composed primarily of hydrated layer-lattice silicates and Fe, Ni-sulfide(s). The most highly ordered layer silicates are serpentine, as indicated by their compositions and basal lattice-fringe spacings (~0.7nm spacing; Fig. 2). The FIB section extracted from the material associated with the ¹⁵N anomaly contains an amorphous carbon grain in addition to the layer silicates. The amorphous carbon grain was subsequently reanalysed on the NanoSIMS to determine its N and C isotopic compositions.

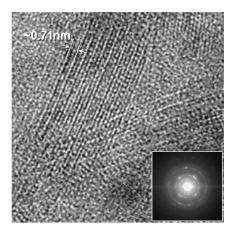


Fig 2: Lattice fringe image of the serpentine within the matrix of L2047 D23. The insert shows the SAED pattern obtained for the silicate phase.

Isotopic Results: The majority of the IDP has $^{14}\text{N}/^{15}\text{N}$ the same as the solar value (272). An ^{15}N -enriched region, ~800 nm in size, was identified with a $^{14}\text{N}/^{15}\text{N}$ ratio of 192 ± 4 (2 σ) compared to 272 ± 2 for the bulk particle (Fig. 3). The N anomaly ($\delta^{15}\text{N}{=}414$) is localized in a discrete grain of amorphous carbon. The O and C isotopic compositions of the IDP are homogeneous throughout at roughly the solar composition.

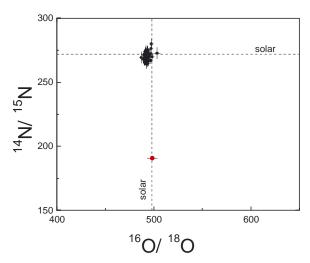


Fig 3: Isotopic composition of the anomalous grain (round red symbol) compared to $3\mu m$ square ROIs. Errors are 2σ .

Discussion: Previous studies of IDPs (e.g. [7]) have made similar observations of a predominantly solar N isotope composition with discrete areas enriched with ¹⁵N. However, in contrast to [7], the ¹⁵N anomaly in L2047 D23 is not associated with any other isotopic anomalies. Extreme ¹⁵N-enrichments have been observed in anhydrous IDPs, presolar

grains, and in meteorites. Presolar grains have 15 N-enrichments of up to 50,000‰ [11]; these anomalies are thought to be from nova explosions [12]. Anhydrous IDPs have 15 N-enrichments of up to $\sim 1300\%$ [7, 9]. Anhydrous IDPs are thought to be from comets based on their porous, fragile microstructures, a lack of significant (aqueous) alteration, and an abundance of Mg-rich silicates common to both the IDPs and some comets [13]. The CN^- complex in Comet Hale-Bopp and Comet LINEAR both give a δ^{15} N of 943‰ [14].

The identification of serpentine as the dominant mineral in L2047 D23 confirms that it belongs to the hydrated chondritic smooth (CS) subset of IDPs. These IDPs may be linked to CM/CI chondrite petrogenesis, underscoring the central role that parent body aqueous activity may have played in the origin of CS IDPs. The preservation of ¹⁵N anomalies in submicron amorphous carbon grains may suggest a late addition of exotic material. It is unlikely that the nonrefractory amorphous carbon could be a carrier for a nucleosynthetic signature. Also, the C isotopic anomalies, which would be expected to accompany a nucleosynthetic origin, are not present, suggesting that chemical fractionation in a cold molecular cloud is the source of the ¹⁵N anomaly. Late in the evolution of molecular clouds, 15N-enriched ammonia, up to ~800‰, may be produced [15]. Whether the anomalous N signature can be transferred from ammonia into the amorphous carbon is unknown. As far as we know, ¹⁵N-enrichments are more extreme in anhydrous than in hydrous IDPs. However, both types of IDPs may have ¹⁵N-enrichments dominated by chemical fractionation in the cold molecular cloud.

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